

Microsecond laser polarimetry for emissivity measurements on liquid metals at high temperatures – application to Niobium¹

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ABSTRACT

Thermophysical data such as enthalpy, specific heat capacity, thermal conductivity, thermal diffusivity or specific electrical resistivity have been measured for about 15 years at the Institut für Experimentalphysik in Graz by using fast pulse heating techniques [1, 2, 3]. Among the properties of most interest is the absolute specimen temperature. Due to the high temperatures achieved and the short duration of the experiments, the temperature measurements must be performed by optical methods. This requires the use of fast pyrometers and knowledge about the behavior of the normal spectral emissivity as a function of temperature. Prior to now, all existing pulse heating systems capable of investigations on liquid samples could only measure the radiance temperature during the experiments. In order to determine true temperature it was necessary to assume that the emissivity was constant over the entire temperature range with a value equal to that at the melting point. This assumption can cause large uncertainties in temperature measurements, especially at elevated temperatures. To avoid these uncertainties, a μ s – Division of Amplitude Photopolarimeter (μ – DOAP) was added to our experiment. The μ – DOAP detects changes of the polarization state of a laser beam ($\lambda = 684.5$ nm) reflected by the sample surface during the pulse – heating experiments. The change in the polarization state is used to determine the index of refraction n and the extinction coefficient, k . This leads to the reflectivity R of the sample and by means of KIRCHHOFF's law for opaque materials the normal spectral emissivity is obtained. The application of simultaneous laser polarimetry, spectral radiometry, and true temperature measurements is demonstrated on liquid niobium.

1. INTRODUCTION

Normal spectral emissivity values in the solid state employ two well – proven methods. In the case of the so – called black body technique, a small hole is machined into the sample. If the ratio of the depth of this hole to the diameter is larger than 10, this hole behaves like a black body source [4] i.e. if one measures the radiation intensity emitted by this hole with a calibrated pyrometer, true temperature can be determined. If the radiance temperature also is measured at the sample surface (if possible near the black body hole in order to avoid uncertainties due to temperature gradients), one obtains normal spectral emissivity at the wavelength of the pyrometers from the ratio of the two intensities with the aid of PLANCK's radiation law.

Another method to determine emissivity in the solid state is the integrating sphere technique [5]. An integrating sphere is positioned close to the surface of the sample (a strip for this case). The sample surface is illuminated with a modulated laser beam and the reflected intensity is collected in the sphere and detected with a photodiode. By means of a lock – in technique, in order to eliminate the background radiation of the hot sample, a signal proportional to the spectral hemispherical reflectivity of the sample is measured. Comparisons with a BaSO₄ reference mounted in the sphere provides the absolute value of reflectance of the sample, and the normal spectral emissivity of the specimen is obtained with the aid of KIRCHOFF's law.

However, these two methods are only applicable to the solid state, since on the one hand the black body source would collapse due to surface tension in the liquid phase, and on the other hand evaporation from the liquid metal would coat and destroy the inner surface of the integrating sphere.

A method of measurement which is applicable to both, the solid and the liquid states is laser polarimetry [6]. With conventional polarimeters the reflected laser beam is often analyzed by

means of rotating polarizer and/or retarder and FOURIER – analyzing the transmitted signal. These devices are not suitable for use in rapid pulse heating experiments due to the rather short experiment durations.

Recently, polarimeters with no moving parts have been developed [4]. The μ s – DOAP is one such device in which the light reflected from the sample is divided into two phase shifted beams by means of a coated beam splitter and each of these two beams is again divided by a GLAN – THOMSON polarizing prism to yield four light fluxes. With a suitable optical design, the polarization states of the four partial beams are independent and the four components of the STOKES vector $\mathbf{S} = (S_0, S_1, S_2, S_3)$ can be calculated from the four measured intensities $\mathbf{I} = (I_0, I_1, I_2, I_3)$ using a 4 x 4 array that represents the calibration matrix, \mathbf{F} , of the DOAP.

Once the STOKES parameters of the reflected light are measured, the normal spectral emissivity (at the wavelength of the laser, in the present case 684.5 nm) can be calculated from the change of the polarization state of the reflected laser beam as described in the subsequent sections.

1. MEASUREMENTS

1.1 Temperature

The optics of a pyrometer views an area of 0.2 mm x 10 mm of the sample surface with a 1:1 magnification onto the rectangular entry slit of an optical waveguide. The interference filter with a center wavelength of 650 nm and a half power bandwidth of 37 nm is in front of the entry slit of the waveguide. The light delivered by the waveguide is detected by a Si - photodiode and amplified with a fast (bandwidth 1 MHz) amplifier. The intensity signal J can be expressed as:

$$J(\lambda, T) = g \cdot \sigma(\lambda) \cdot \tau(\lambda) \cdot \varepsilon(\lambda, T) \cdot \frac{c_1}{\lambda^5 \left[e^{\frac{c_2}{\lambda \cdot T}} - 1 \right]} \quad (1)$$

where the symbols are given as g , geometry factor; σ , sensitive of the electronics and of the diode; λ , wavelength; τ , transmission of the optics and of the optical waveguide; ε , normal spectral emissivity, c_1 and c_2 , first and second PLANCK's constant; T , temperature

For a calibrated pyrometer the following relation between the detected intensity signal and radiance temperature is valid:

$$T_{Rad} = \frac{c_2}{\lambda \cdot \ln \left(\frac{K}{J} + 1 \right)} \quad (2)$$

where: K , calibration factor; J , pyrometer signal

The unknown quantities g , σ and τ are combined into the calibration factor K , which only requires a linear amplifier, and is independent of temperature. From PLANCK's radiation law the relation between radiance temperature and true temperature is given by:

$$T_{true} = \frac{c_2}{\lambda} \cdot \frac{1}{\ln \left[\varepsilon \left(e^{\frac{c_2}{\lambda \cdot T_{Rad}}} - 1 \right) + 1 \right]} \quad (3)$$

Prior to this work, all experiments involving rapid pulse heating into the liquid state assumed a constant emissivity value in the liquid phase and equal to the value deduced from the melting transition [7, 8]. In the following section, we describe the construction and operating

principles of a laser polarimeter (DOAP) system that makes direct measurements of the normal spectral emissivity in the liquid state during rapid pulse heating experiments.

1.2 Normal spectral emissivity

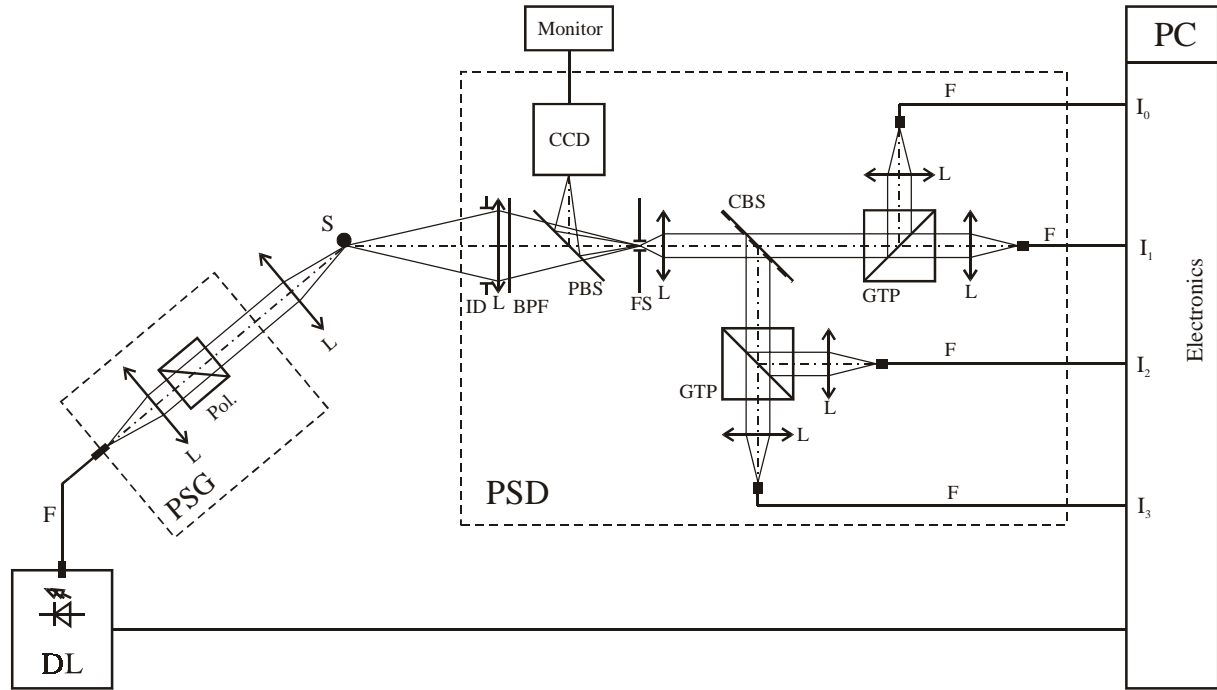


Fig. 1: Schematic drawing of the μs – DOAP. CCD, CCD camera with monitor; PSG polarization state generator; PSD, polarization state detector; DL, diode laser; Pol., polarizer; L, lenses; S, sample; BPF, band pass filter; PBS, pellicle beam splitter; CBS, coated beam splitter; GTP, GLAN – THOMSON prism; F, fiber; ID, iris diaphragm; FS, field stop; $I_{0,1,2,3}$, four intensities pictured to diodes; PC, personal computer.

Figure 1 shows the setup of the μs - DOAP system. On account of the rapid changes in the current through the sample which occurs during a pulse heating experiment, both, the source of light and the electronics are placed in a shielded room. The signals from the experiment are transferred by means of optical waveguides into the shielded room. A semiconductor laser diode, which produces radiation with a wavelength of 684.5 nm serves as the light source.

This light is amplitude modulated at a frequency of 8 MHz. The light from the optical waveguide is delivered to the polarization state generator (PSG), which consists of a pair of motorized rotators that hold a linear polarizer and a quarter wave retarder such that any desired incident polarization state can be generated and delivered to the specimen. The polarized and modulated laser beam is focused onto the sample, and the reflected light collected by the Polarization State Detector (PSD). The PSD divides the reflected laser beam into four beams, which are collected by optical waveguides and delivered to four Si – photodiodes. The detector output is demodulated using a lock – in technique with an output bandwidth approaching 2 MHz. From the four obtained signals $\mathbf{I} = (I_0, I_1, I_2, I_3)$ one receives via the calibration matrix \mathbf{F}^{-1} the four components of the STOKES vector $\mathbf{S} = (S_0, S_1, S_2, S_3)$ of the reflected laser beam from which the specimen properties can be derived. The matrix \mathbf{F} is determined separately from a calibration procedure [9].

$$\mathbf{S} = \mathbf{F}^{-1} \cdot \mathbf{I} \quad (4)$$

If the incident polarization state is set to $+45^\circ$ (linear), and the reflected STOKES parameters (S_0, S_1, S_2, S_3) , are measured, the ellipsometric parameters, ψ and Δ , are given by:

$$\psi = \frac{1}{2} \tan^{-1} \left[\frac{\sqrt{S_3^2 + S_2^2}}{-S_1} \right] \quad (5)$$

$$\Delta = \tan^{-1} \left[\frac{-S_3}{S_2} \right] \quad (6)$$

The values of ψ , Δ and the incidence angle Θ yield the refractive index n , and the extinction coefficient k of the reflecting materials using the following equation:

$$n - i k = n_0 \tan(\Theta) \left[1 - \frac{4 \rho \sin^2(\Theta)}{(1 + \rho)^2} \right]^{\frac{1}{2}} \quad (7)$$

where $\rho = \tan(\psi) \exp(i\Delta)$ and n_0 is the refractive index of the transparent ambient medium.

Once n and k have been determined, the normal spectral reflectivity R , is calculated from the following equation:

$$R = \frac{(n - n_0)^2 + k^2}{(n + n_0)^2 + k^2} \quad (8)$$

For opaque materials the normal spectral emissivity $\varepsilon(\lambda)$ is then determined from KIRCHOFF's law,

$$\varepsilon(\lambda) = 1 - R(\lambda). \quad (9)$$

3. RESULTS

Figure 2 presents the time dependent behavior of the four intensity signals obtained by the DOAP system and the corresponding radiance temperature of a single experiment on Niobium. One can see that all four DOAP signals have a strong rise in intensity at the start of melting. This can be explained by the reduction in surface roughness and the formation of a specular liquid surface, giving rise to an increase in reflectivity.

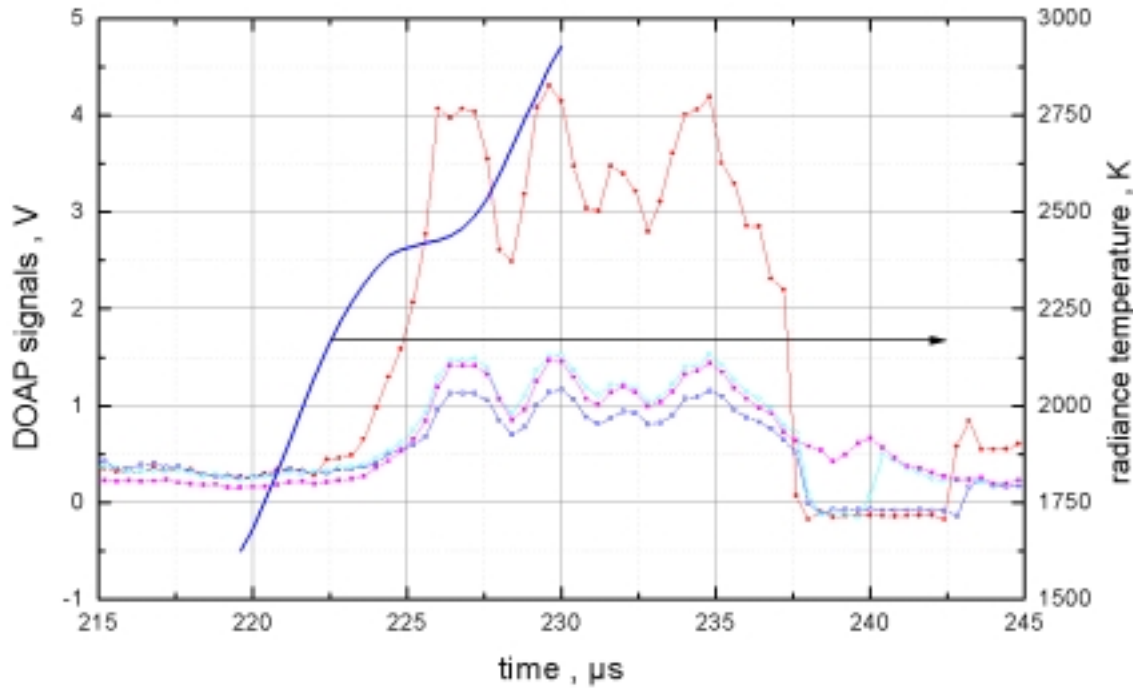


Fig. 2: Four intensity signals of the DOAP as well as radiance temperature versus time for an experiment on Niobium.

For both, the solid and the liquid state, the noise of the demodulated signals should have the same value, but it is observed that the signals in the liquid state are associated with a substantially greater noise. The noise appears to be correlated among the four channels and is associated with the non – gaussian shape of the laser beam. As stated previously, the laser light source is delivered to the specimen using an optical fiber. Due to the small surface of the wire (\varnothing 0.5 mm) only a small amount of intensity is reflected, therefore a multimode fiber optic waveguide is used. In order to get more reflected intensity in addition, the laser beam is focused onto the sample surface. Multimode fibers are always associated with a strong non – gaussian profile (see Fig. 5). This arrangement is very sensitive to specimen motion. Since the sample expands significantly during the experiment, especially in the liquid state, it

is believed that this correlated noise arises due to reflection by the wire from different points of varying intensity across the multimode laser beam profile.

Figure 3 depicts normal spectral emissivity at 684.5 nm versus time for the same experiment. The start of the experiment at room temperature is at about 204 μs , the polarimeter signals and thus calculated emissivity values are strongly disturbed by the firing of the ignitron, which is used as a fast current switch. The liquid phase begins around 225 μs and ends around 235 μs . Three peaks can be observed in this time interval which match exactly with the correlated noise peaks discussed previously in Fig. 2. A strong peak at 234 μs occurs, which corresponds to the collapse of the the four signals (see Fig. 2). The behaviour of the emissivity in the solid state is quite noisy but the emissivity in the solid is not very repeatable since the emissivity is influenced mainly by roughness of the surface. To obtain reproducible values for the solid state, one must pre – heat the sample several times close to melting to smooth the surface and to eliminate containments [10, 11]. However, this was not the aim of these investigations, whose focus was to obtain data in the liquid phase.

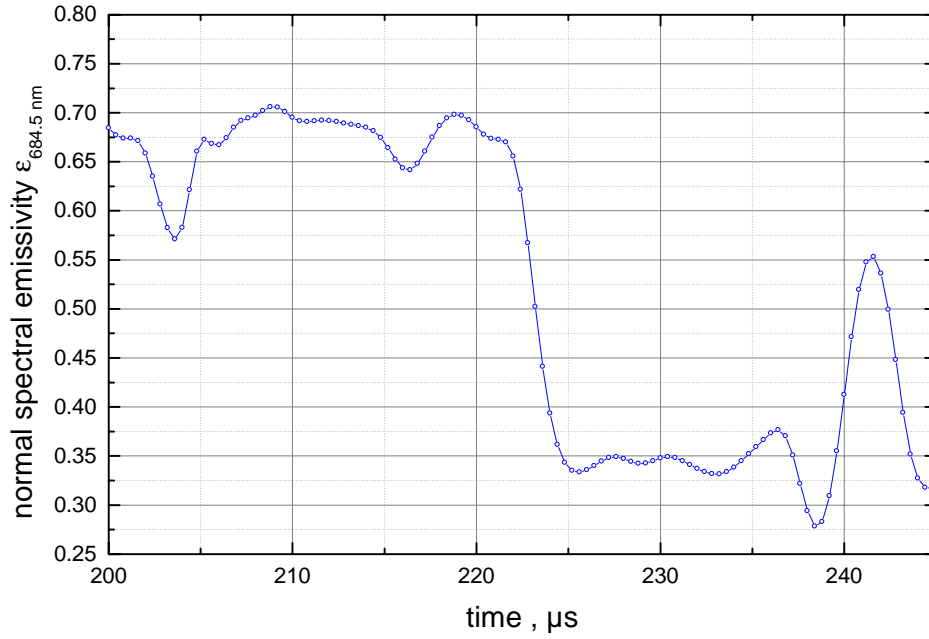


Fig. 3: Emissivity at 684.5 nm for a single experiment on a niobium sample versus time.

The dependence of normal spectral emissivity at 684.5 nm on radiance temperature is presented in Fig. 4. Since the amplifier of our pyrometer had an upper radiance temperature limit of about 3000 K, measured values are only available up to this temperature. To obtain values at higher temperatures one would have to decrease the radiance from the sample using neutral density filters or by varying of the amplification.

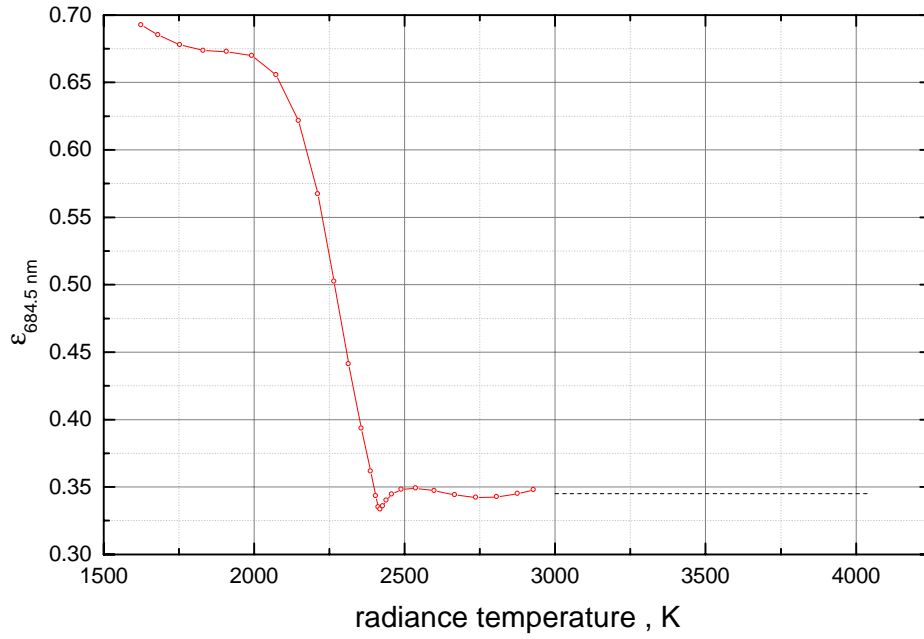


Fig. 4: Emissivity at 684.5 nm as function of time for niobium. The full line indicates measured values up to 3000 K, the dashed line shows an extrapolation up to 4000 K.

In Figure 4 the spectral emissivity drops just before melting from a value of 0.7 to a value of 0.33 at the onset of melting. At the end of melting the emissivity increases slightly to 0.345 and remains more or less constant up to 3000 K (radiance temperature).

4. CONCLUSIONS

If we assume a linear increase in the temperature with time, we obtain a radiance temperature of (4000 ± 50) K at the point of the collapse of the four intensity signals (at $236,5 \mu\text{s}$). With an emissivity value of 0.345 measured by the DOAP, we obtain a true temperature of (5000 ± 70) K. This value is in good agreement with the boiling point of 5015 K of niobium [12]. This would indicate that the DOAP system provides emissivity values for the whole liquid state for niobium.

The beam shape of the laser, after the optical waveguide and the PSG – module is presented in Fig. 5. One can see, that individual fibers of the waveguide show different polarization states. Depending on how the polarization state of the corresponding fiber is, one receives after the linear polarizer and the quarter wave retarder of the PSG - module a corresponding intensity, what leads to the shown beam shape. To avoid this problem, we plan to eliminate the waveguide and to position the laser in a shielded box in front of the PSG – module to obtain a perfect gaussian beam profile and thus avoiding the strong variations of intensity in the liquid phase.

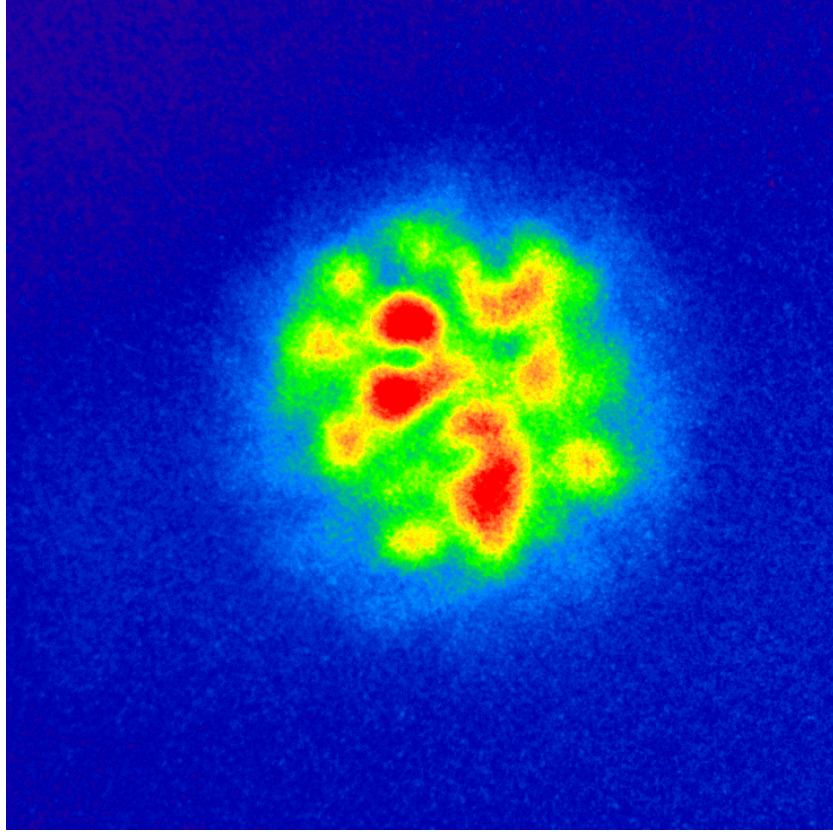


Fig. 5: Beamprofile of the laserbeam after PSG – module, detected by a 12 bit intensified CCD – camera (DiCAM Pro, PCO, Regensburg, GERMANY).

5. UNCERTAINTIES

The absolute accuracy of the DOAP measurements of normal spectral emissivity is estimated to be ± 0.015 or about $\pm 5\%$ for the present measurements. This uncertainty is greater than that which would be predicted from the polarization state measuring ability of the instrument, but consistent with the known properties of niobium at the melting point [4]. Independent measurements on reference surfaces showed that the index of refraction, n , on a BK7 glass surface was measured to an accuracy of 0.5% or better. The precision of the measurements is also somewhat larger, approximately, ± 0.01 , due to the fact that measurements on the liquid state are considerably more difficult due to the speed, high background radiances, and the rapid changes in the intensity which occur due to the non – gaussian beam profiles.

6. ACKNOWLEDGEMENT

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